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Influence of random roughness on the adhesion between metal surfaces due to capillary condensation

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The capillary force was measured by atomic force microscopy between a gold coated sphere mounted on a cantilever and gold surfaces with different roughnesses. For smooth surfaces the capillary adhesive force surpasses in magnitude any dispersion, e.g., van der Waals/Casimir and/or electrostatic forces. A substantial decrease in the capillary force was observed by increasing the roughness amplitude a few nanometers in the range of 1–10 nm. From these measurements two limits can be defined: a smooth limit where a closely macroscopic size contact surface interacts through the capillary force and a rough limit where only a few asperities give a capillary contribution. © 2007 American Institute of Physics. [DOI: 10.1063/1.2768919]

Adhesion is an important phenomenon in emerging technologies related to microelectromechanical systems (MEMS), where preventing sticking of device components is a major challenge. In general, MEMS are sensitive to adhesion as a result of the large surface-to-volume ratio, of the micro—to submicrometer separations, and of the highly compliant components. Dispersive surface forces (e.g., van der Waals/Casimir), which are relatively well understood,^{1,2} contribute to adhesion and play a major role during the fabrication and operation of MEMS.^{3–8} However, complications arise when dealing with real systems due to surface roughness and capillarity driven forces.

Capillary condensation occurs into small cracks and pores by wetting of liquids that originate from water vapor. As a result, when the planar surfaces of MEM microswitches come in close proximity, attractive capillary forces can start acting on them. The latter occurs if the surface separation becomes $\approx 2R_k \cos(\theta)$, with θ as the contact angle.⁹ At equilibrium, the Kelvin radius R_k (which is related to the meniscus height) is related to the relative vapor pressure P/P_{sat} at temperature T by the Kelvin equation $R_k = -(\gamma V_m / RT) \times [\log(P/P_{\text{sat}})]^{-1}$.^{1,9} γ is the liquid vapor pressure (e.g., $\gamma = 73 \text{ mJ/m}^2$ for water), and V_m is the molar volume of the liquid. However, the process by which asperity phenomena bridge the length scale causing adhesion of micro-macroscopic interfaces is still unknown.

Recently, it was shown that below a threshold of relative humidity (RH) P/P_{sat} , the adhesion was mainly due to van der Waals forces across extensive noncontacting areas between SiO₂ surfaces, while above this threshold of RH, capillary condensation was the predominant term.¹⁰ The adhesion energy was found to vary from 10^{-3} to 10^2 mJ/m^2 , with values highly depending on the surface roughness (the plate roughness varied from 2 to 10 nm, while the cantilever roughness was not mentioned) and high RH (threshold $\geq 70\%$ depending on roughness). In former studies of the roughness influence on adhesive forces, either surface with a

relatively high roughness or a low amount of samples was used.¹¹

Therefore, in this letter, we will investigate the adhesion force compared to other dispersion forces (van der Waals/Casimir) between gold (Au) surfaces as a function of the nanoscale roughness amplitude at a relative humidity below 70% and the lateral correlation length. For this purpose the roughness of the plate was systematically and carefully increased from atomically flat annealed films on mica up to approximately 10 nm rms roughness amplitude for vapor deposited Au films.

The piconewton atomic force microscopy (AFM) from Veeco was used for the force measurements, where creep and hysteresis are eliminated with the closed loop scanner (having a noise level below $<0.5 \text{ nm}$ and a nonlinearity of 0.1% over a $20 \mu\text{m}$ range). Adhesive forces were measured by retracting a sphere attached on the cantilever from the surface. The spring constant is measured from eight electrostatic force curves with applied voltages between ± 3 and $\pm 4.5 \text{ V}$ (error of 1 mV), at a distance above $1 \mu\text{m}$, yielding $k = 5 \text{ N/m}$ (error less $<10\%$). Polystyrene spheres $100 \mu\text{m}$ in diameter, $2R_s$, with 1.5% deviation from sphericity) were used. AFM was employed to determine the roughness of the sphere and yielded a 1.2 nm rms roughness amplitude (before coating) over a $25 \mu\text{m}^2$ area. The spheres were coated with 100 nm Au in an evaporator (at 10^{-6} mbar base pressure) and at a deposition rate of 0.6 nm/s yielding surfaces with 1.8 nm rms roughness. The force measurements were repeated with different spheres having the same rms roughness amplitude and cantilevers. After the force measurement, inspection with scanning electron microscopy did not reveal any damage to the Au layer on the sphere. In addition, successive AFM scans (Fig. 1) revealed the typical roughness of the Au film on the sphere, indicating that the Au layer remained unmodified by the measurement process. The contact potential has been eliminated within a 20 mV error, which resulted in electrostatic forces much smaller than the van der Waals force at the closest separation.

In this setup, the sphere surface sets a lower limit on the combined smoothness of the adhering objects. Therefore, it is important to investigate whether the perfectly smooth limit

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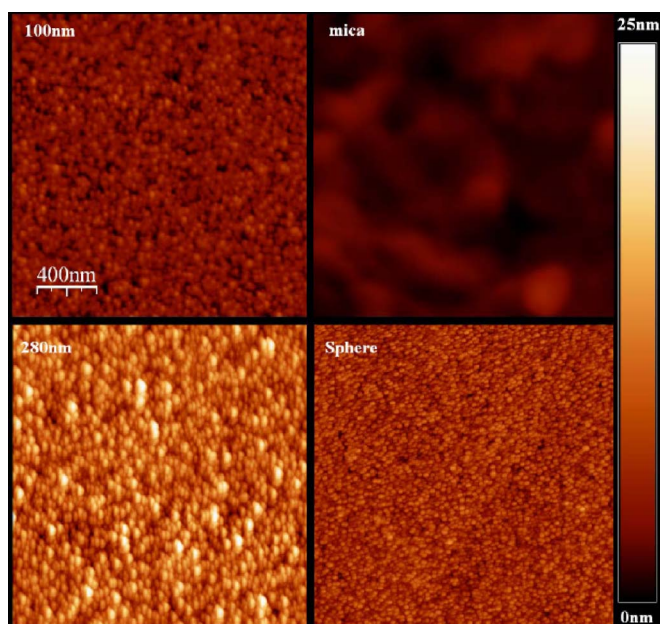


FIG. 1. (Color online) Surface scans of the 100 nm gold on mica, 280 nm films, and the coated sphere surface; the scan size is $2\ \mu\text{m}$ and the color range corresponds to 25 nm z range, where lighter shades mean higher areas.

is reached. Au films from 100 up to 1200 nm were grown on silicon wafers and those at 150 nm on mica. The latter sample was annealed extensively to obtain very smooth surfaces (see Fig. 2). The measured roughness varied from 0.8 nm rms for the Au/mica and up to 1.1–9 nm for the Au/SiO₂ films. Note, however, that the Au/mica film is very smooth locally, indicated by the large correlation length $\xi \approx 100$ nm and its height variations of atomic steps in size. The correlation length of the Au/SiO₂ films is much smaller within $\xi \approx 15$ –40 nm. For gauging the effect of correlation length on the adhesive force, another film (120 nm thick) was made on a smoother wafer, which resulted in a rms roughness of 1.5 nm but a correlation length about three times larger than that of the 100 nm thick Au/SiO₂ film (Fig. 2).

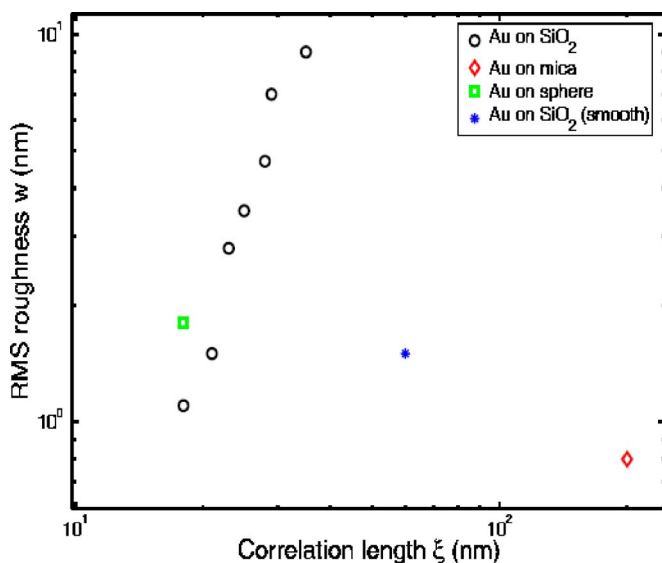


FIG. 2. (Color online) Plots of the roughness amplitude w vs lateral correlation length ξ for the gold grown on SiO₂, mica, and sphere.

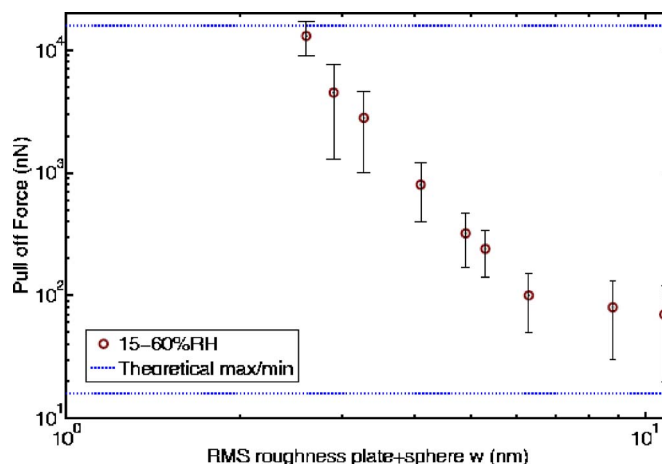


FIG. 3. (Color online) Adhesion force vs rms roughness of the sphere and plate added together. The upper and lower horizontal lines represent the theoretical values for capillary interaction, in the case of a perfectly smooth sphere with a perfectly smooth plate and in the case of a single asperity with a correlation length of 50 nm.

Figure 3 shows that the adhesion force remains very large for weakly rough surfaces, and follows a rapid decrease with more than two orders of magnitude for a variation of the rms roughness amplitude by a few nanometers in the range 1–10 nm. This range of roughness is typical for fabricated nanoelectromechanical systems (NEMS) surfaces,¹⁰ and, therefore, sets the limit over which capillary forces are of importance in nanodevices covered with metallic layers. The results for the 120 nm film (1.5 nm rms roughness) with a large correlation length of 60 nm did not yield higher forces than the 100 nm thick film with a correlation length of 20 nm (also 1.5 nm rms). Therefore, we can conclude that the rms roughness gives the predominant effect on the adhesive force due to capillary condensation.

From the roughness scans on different locations on the surface, the variation in the rms roughness is ~ 0.1 nm, while the standard deviation in adhesion force was 30%–70% (Fig. 3) for the rougher films when measuring at 50–100 different surface locations. The latter is larger than the error due to the calibration of the cantilever spring constant and originates from a set of different reasons. First it is important to realize that the capillary force is extremely sensitive to nanoscale roughness, where it falls two orders of magnitude with increasing rms roughness in the range 1–10 nm (Fig. 3). Therefore, it is not surprising to obtain a force a few times smaller on the same plate if there are some local protruding peaks that may also deform. The latter was visible in some films where the force gradually increased up to five times after immediate successive measurements at the same location. All of these effects were, however, not seen for the Au/mica film, which is locally much smoother and does not have any peaks that deform easily. In contrast, high peaks were abundant in the other Au films and the correlation length was found to be small. The load was kept low during the approach to avoid significant influence on the adhesion force beyond the stated error bars. Another explanation for the force deviation originates from scanning tunneling microscopy (STM) studies, which indicated that below 60% RH the water film on Au is not closed but droplet formation takes place (even for RH as low as 15%).¹² The average height of droplets ~ 5 –10 nm did not decrease much with

lower RH, while the amount and size of droplets was decreased.¹²

The total adhesion force can be divided into a capillary force and an interfacial tension force due to surface tension acting tangentially to the interface along the contact line with the solid body.¹¹ For the relatively smooth films, the theory prediction for flat surfaces gives an approximate upper limit for the force $F_{\text{up}} = 4\pi\gamma R_s \cos(\theta)$ Ref. 11 ($F_{\text{up}} = S\Delta\sigma$, with $S = 4\pi R_s R_k \cos(\theta)$, the meniscus surface area, and $\Delta\sigma = \gamma/R_k$, the Laplace pressure, while ignoring contributions from surface tension). For the contact angle of water onto Au surfaces of 70° Ref. 12 we obtain for $R_s = 50 \mu\text{m}$, $F_{\text{up}} = 1.5 \times 10^4 \text{ nN}$. With increasing roughness, surfaces can be more hydrophobic,¹³ leading to reduced capillary force. In addition, the contact area between sphere and plate is highly reduced. If we consider the lower limit for the force in terms of contact onto a single asperity,¹⁴ where the capillary meniscus is formed only between this asperity and the surface of the sphere, we obtain a force of magnitude $F_{\text{low}} = 4\pi\gamma\xi \cos(\theta) = 15 \text{ nN}$ (where we used as an effective asperity radius the roughness correlation length ξ). From Fig. 3 it appears that the smooth limit is reached for the Au/mica film. For the roughest films the values found are up to ten times higher than that of a single asperity, indicating a capillary interaction of a multitude of asperities.

When the RH is changed from 15% up to 60%, no significant change in capillary adhesion was observed, although we do not rule out any change because the standard deviation in the measurement is rather large. Again, the STM analysis of the incomplete wetting of the water layer mentioned earlier gives a good explanation for this behavior.¹¹ In Ref. 10 it was reported that for very high RH close to 100%, corresponding to large kelvin condensation length, the capillary force reaches a maximum value even for the roughest surfaces. This is not surprising as at high RH the water layer on the film increases rapidly, even surpassing the highest roughness peaks. The capillary force overshadows any other force (van der Waals/similarity or electrostatic) as being 100–1000 times stronger for smooth surfaces. For rough surfaces the Casimir force becomes comparable within an order of magnitude to the capillary force. The Casimir force between plate and sphere is roughly 5 nN for the rough surfaces in the range of contact, whereas similar sized capillary forces

(15 nN) are produced by single asperities with a lateral size of about 30 nm.

In conclusion, we have shown that for relatively low humidity capillary forces are present in the case of smooth surfaces and surpass in magnitude any dispersion and electrostatic forces. In addition, an enormous decrease in the capillary force was observed by increasing the roughness amplitude a few nanometers in the range $\sim 1\text{--}10 \text{ nm}$. Considering the rapid fall-off in the capillary force and the two limits (a smooth limit where the whole surface contribute to the capillary force, and a rough limit where only a single or a few asperities contribute), the crossover regime might, in turn, depend on the contact angle and any lateral roughness features. Both could be an interesting direction for further study of this phenomenon in the design of MEMS/NEMS if stiction poses a problem.

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